



METHOD FOR IMPROVING THE RELIABILITY OF BRITTLE MATERIALS THROUGH THE CREATION OF A THRESHOLD STRENGTH

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BACKGROUND OF THE INVENTION

Field of Invention

[0002] This discovery concerns the improvement of the reliability and damage tolerance of brittle materials through the use of novel crack arresting architectures composed of compressive layers specifically placed throughout the body that prevent failure from occurring until a specific, predetermined threshold strength is reached.

Description of Related Art

[0003] The strength of most common brittle materials is not deterministic, i.e. single-valued, due to the presence of an unknown distribution of strength-limiting flaws inadvertently introduced during processing and surface machining [1,2]. As a result, the strength of brittle materials must generally be described by a statistical distribution of strengths with associated probabilities of failure at each of those strengths. Failure from these types of flaws is generally not an issue in ductile materials because they exhibit plastic deformation that desensitizes the relation between small flaws and strength. Plastic deformation also absorbs work from the loading system to increase the material's resistance to the extension of large cracks. However, the lack of plastic deformation in brittle materials causes their strength to be inversely dependent on the size of very small cracks, which generally cannot be detected except by failure itself.

[0004] Consequently, design with brittle materials generally becomes a practice of defining acceptable levels of reliability. Designers must not only make

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accommodations for probabilistic definitions of the strength and the finite probability of failure at any applied stress, but they must also be further concerned with the fact that, once in service, seemingly insignificant and sometimes undetectable damage could be incurred that would drastically reduce the load carrying ability of the material. This lack of reliability is one of the major reasons why brittle materials have not been more widely used, despite the potential they offer for substantial performance enhancements in a wide variety of applications.

[0005] One method for improving the reliability of components made from brittle materials has been through the use of proof testing. The proof test is designed to emulate the thermomechanical stresses experienced by the component in severe service and defines a threshold stress below which components are eliminated by failure prior to service. However, given its destructive nature, proof testing is generally only used when performance needs outweigh consumer price sensitivity. In ceramics, another approach to ensuring reliability is by eliminating heterogeneities that give rise to flaws, such as inclusions and agglomerates, from the ceramic powder. One method to remove heterogeneities greater than a given size is to disperse the powder in a liquid and pass the slurry through a filter [1]. If heterogeneities are not reintroduced in subsequent processing steps, and surface cracks introduced during machining are not a critical issue, filtration determines a threshold strength by defining the largest flaw that can be present in the powder and thus, within the finished ceramic component [3]. However, neither of these techniques mitigates the detrimental effect of service-related damage.

[0006] Recently, another method for improving reliability through the use of residual, compressive stresses that have their maxima located some specific distance beneath the surface of the material was proposed by Green et al [4]. The authors suggested that the unique compressive stress profiles they developed would arrest surface cracks and lead to higher failure stresses and improved reliability through reduced strength variability. However, compressive stresses, either at or just beneath the surface, will not effectively hinder internal cracks and flaws, nor can they produce a threshold strength; thus high reliability is still not ensured. As shown below, a threshold strength can only arise when compressive layers are placed on the surface

and throughout the body to interact with both surface cracks and internal cracks and flaws.

SUMMARY OF THE INVENTION

[0007] The present invention provides a new method for fabricating reliable, damage-tolerant brittle materials. By incorporating layers of one material under residual compression on the surface and throughout the bulk of one or more other materials, a composite is formed in which the propagation of otherwise catastrophic cracking is arrested. The residual compression within these layers acts to reduce the stress intensity of the cracks, thereby causing them to arrest until further loading is provided. This highly desirable stable, subcritical crack growth mode persists with increased loading until the applied stress is large enough to drive the crack completely through compressive region, after which failure occurs.

[0008] The exact level of stress needed to cause failure is dictated by the architectural design of the compressive layers such that the material can be designed to have any minimum strength desired, within the limits of the materials system used. This results in a truncation of the strength distribution, such that there is virtually zero probability of failure below this minimum value, i.e. the material possesses a *threshold strength*. Consequently, sensitivity to flaws that would ordinarily cause catastrophic failure at stresses below the threshold strength is eliminated. Furthermore, the potential exists for the complete elimination of the strength variability, hence improving reliability, through the creation of nearly deterministic, i.e. single-valued, strengths by increasing the threshold strength above the stresses at which failure normally initiates from intrinsic flaws.

[0009] The potential this invention offers for the implementation of brittle materials in high-performance structural applications ranging from high-temperature gas turbine engines to biomedical prosthetic implants is significant. Elimination of the need for designers to rely on probabilistic strength definitions and acceptable failure probabilities will allow for design using conventional engineering methodologies, thereby facilitating the introduction of these materials.

[0010] These and other features, aspects, and advantages of the present invention will become better understood with regard to the following detailed description, claims, and accompanying drawings.

BRIEF DESCRIPTION OF THE DRAWINGS

[0011] **Figure 1** is a schematic representation of the superimposed stress fields used to determine the stress intensity of the arrested crack;

[0012] **Figure 2** shows the results of the optimization analysis as a plot of t_2/t_1 as a function of the independent variables;

[0013] **Figure 3** is a plot of the failure strength of the laminated and monolithic specimens with varying starting flaw sizes;

[0014] **Figure 4** is a scanning electron micrograph of the fracture surface of one of the laminate specimens;

[0015] **Figure 5** is a plot of the threshold strength vs. magnitude of residual compression within the compressive layers for two different laminate architectures;

[0016] **Figure 6** is a plot of the threshold strength vs. compressive layer thickness for two different laminate architectures;

[0017] **Figure 7** is a plot of the threshold strength vs. tensile layer thickness for two different laminate architectures;

[0018] **Figure 8** is an optical micrograph of replicas made of a crack propagating across the tensile surface of a non-edge cracked laminate;

[0019] **Figure 9** is a plot of the stress intensity vs. the crack length for a crack propagating through a non-edge cracked compressive layer;

[0020] **Figure 10** is an optical micrograph of replicas made of a crack propagating across the tensile surface of an edge cracked laminate;

[0021] **Figure 11** is a plot of the stress intensity vs. the crack length for a crack propagating through an edge cracked compressive layer;

[0022] **Figure 12** is a scanning electron micrograph of the fracture surface of a specimen that exhibited edge cracking;

[0023] **Figure 13** is an optical micrograph showing the bifurcation of the crack within the bulk of an edge cracked specimen, beneath the penetration depth of the edge crack;

[0024] **Figure 14** is a plot of the stress intensity of a crack propagating through the compressive layer as a function of distance showing the effect of modulus mismatch;

[0025] **Figure 15** is a plot of the threshold strength vs. amount of $m\text{-ZrO}_2$ for Al_2O_3 laminates with Al_2O_3 / $m\text{-ZrO}_2$ compressive layers;

[0026] **Figure 16** is a schematic diagram of the composite architecture needed to produce compressive layers within a body formed of hexagonal prisms of one or more materials, separated and bonded together with compressive layers formed of another material; and

[0027] **Figure 17** is a schematic diagram of the composite architecture needed to produce compressive layers within a body formed of polyhedra of one or more materials, separated and bonded together with compressive layers formed of another material.

DETAILED DESCRIPTION

[0028] The hypothesis that multiple, thin compressive layers could lead to a threshold strength in brittle materials had its genesis in an inadvertent observation made by one of us (Antonio Javier Sanchez Herencia) in which a crack was observed to initiate and arrest between two compressive layers during experiments to further understand the phenomena of crack bifurcation, that is, the 90° change in the direction of a crack as it enters and extends along the center line of a compressive layer [5-7]. This observation initiated a fracture mechanics analysis to determine the conditions for crack arrest and subsequent failure, and subsequent experiments to test the analysis [8].

Mechanical Analysis of the Arrest Phenomena and Development of the Threshold Strength Model

[0029] A residual, biaxial, compressive stress arises within layers of thickness t_1 , either surface or internal, when they are compressed relative to a second set of alternating layers of thickness, t_2 . This compression can arise due to a compressive strain mismatch, ϵ , caused by either a mismatch in the thermal expansion coefficients of the laminae, or by a volume change of either of the laminae through a crystallographic phase transformation or formation of a chemical reaction product. For the specific case of a laminated plate composed of compressive layers (t_1), alternated between tensile layers (t_2), the biaxial stresses in both layers are given by [9]

$$(1) \quad \sigma_1 = \epsilon E_1' \left(1 + \frac{t_1 E_1'}{t_2 E_2'} \right)^{-1} \quad \text{and} \quad \sigma_2 = -\sigma_1 \frac{t_1}{t_2}$$

where $E_1' = E_1 / (1 - \nu_1)$, E is Young's modulus and ν is Poisson's ratio. Inspection of the two relations shows that thin compressive layers are desired because when $t_1/t_2 \rightarrow 0$ the compressive stress is maximized while the tensile stress diminishes to zero in the thicker layers.

[0001] The analysis of the observed arrest phenomena assumes that a pre-existing crack of length $2a$ spans the thick layer (t_2), sandwiched by the compressive, thin layers of thickness t_1 , as shown on the left side of Fig. 1. The magnitude of the biaxial, residual compressive stress within the thin layers is given by σ_c , and the opposing residual tensile stress within the thick layer is given by σ_t . The analysis determines the stress intensity factor for a crack of length $2a$ when it extends into the compressive layers ($t_2 \leq 2a \leq t_2 + 2t_1$), under an applied stress, σ_a , parallel to the layers. The stress intensity factor is used to determine the applied stress, σ_{thr} , needed to extend the crack through the compressive layers to produce catastrophic failure.

[0031] The stress intensity factor, K , is determined by superimposing the two stress fields shown on the right hand side of Fig. 1, each applied to the same slit crack of length $2a$, and each with its own, known stress intensity factor. The first is a tensile stress of magnitude $\sigma_a - \sigma_c$ applied to a cracked specimen that does not contain

residual stresses. The stress intensity factor for this stress is given by the first term on the right hand side of Eq. 2A. The second is a tensile stress of magnitude $\sigma_c + \sigma_t$, which is only applied across the thick layer, the portion of crack defined by t_2 . Its stress intensity factor is given by the second term on the right side of Eq. 2A [10]. The two superimposed stress fields sum to that shown on the left hand side of Fig. 1. The stress intensity factor for the two superimposed stress fields is thus given by

$$K = (\sigma_a - \sigma_c)\sqrt{\pi a} + (\sigma_c + \sigma_t)\sqrt{\pi a} \left[\frac{2}{\pi} \sin^{-1} \left(\frac{t_2}{2a} \right) \right] \quad (2A)$$

Substituting $\sigma_t = \sigma_c t_1/t_2$ (from Eq. 1) and rearranging, Eq. 2A better expresses the physical significance of the compressive layers:

$$K = \sigma_a\sqrt{\pi a} + \sigma_c\sqrt{\pi a} \left[\left(1 + \frac{t_1}{t_2} \right) \frac{2}{\pi} \sin^{-1} \left(\frac{t_2}{2a} \right) - 1 \right] \quad (2B)$$

The first term in Eq. 2B is the well know stress intensity factor for a slit crack in an applied tensile field. The second term is always negative and thus reduces the stress intensity factor when the crack extends into the compressive layers. Thus, the compressive layers increase the material's resistance to crack extension.

[0032] Because K decreases as the crack extends into the compressive layers, the maximum stress needed to cause the crack to 'break' through the compressive layers occurs when $2a = t_2 + 2t_1$ and $K = K_c$, the critical stress intensity factor of the thin layer material, a property that describes its intrinsic resistance to crack extension. Substituting these values into Eq. 2B and rearranging, the largest stress needed to extend the crack through the compressive layers is given by

$$\sigma_{thr} = \frac{K_c}{\sqrt{\pi \frac{t_2}{2} \left(1 + \frac{2t_1}{t_2} \right)}} + \sigma_c \left[1 - \left(1 + \frac{t_1}{t_2} \right) \frac{2}{\pi} \sin^{-1} \left(\frac{1}{1 + \frac{2t_1}{t_2}} \right) \right] \quad (3)$$

Equation 3 shows that σ_{thr} increases with the fracture toughness of the thin layer material, K_c , the magnitude of the compressive stress, σ_c , and the thickness of the compressive layer, t_1 . One can also show that if the initial crack length in the thick

layer is $< t_2$, and the stress needed to extend it is $< \sigma_{thr}$, the crack will be arrested by the compressive layers. However, if the crack is very small and extends at a stress $> \sigma_{thr}$, it will extend through the compressive layers to cause catastrophic failure without being arrested. Thus, Eq. 3 defines a threshold stress, σ_{thr} , below which the laminar body cannot fail when the tensile stress is applied parallel to the layers. This prediction has significant implications in that it offers the opportunity to design structural components with the knowledge that the component will not fail below the specified threshold stress.

[0033] It has been shown [11] that the conditions that optimize the threshold strength could be determined by differentiating Eq. 3 with respect to the dependent variable, t_2/t_1 . Figure 2 shows the results of this optimization analysis, which plots t_2/t_1 as a function of the independent variables. Optimum threshold strengths are obtained by choosing values for the independent variables that can be practically achieved through material choice and processing constraints, and then determining the dependent variable, t_2/t_1 using Fig. 2. Inspection of Eq. 3 shows that minimization of the layer thicknesses is desirable, as it will lead to higher threshold strengths. However, current processing technology limits the minimum thickness achievable to $\sim 5 \mu\text{m}$; this thus serves as the constraint for the optimization. For the given Al_2O_3 / mullite system with $5 \mu\text{m}$ thick compressive layers, Fig. 2 shows that threshold strength will be optimized at a t_2/t_1 ratio of ~ 1.6 , thus $t_2 = 7.95 \mu\text{m}$ and $\sigma_c = 1085 \text{ MPa}$. Substituting these values into Eq. 3, one finds that a threshold strength of 945 GPa is possible in this particular system, which is nearly as large as the compressive stress, despite the fact that the ratio $t_2/t_1 = 1.6$ produces large tensile stresses in the thicker layers separating the compressive layers.

[0034] While this is an impressive result, even higher threshold strengths far in excess of the residual compression may be possible in systems with higher toughness compressive layers. Application of Fig. 2 to a laminate system composed of silicon carbide (SiC) tensile layers and silicon nitride (Si_3N_4 , $K_c = 8 \text{ MPa}\cdot\text{m}^{1/2}$) compressive layers, with the compressive layers again constrained to $5 \mu\text{m}$, yields an estimated threshold strength in excess of 2 GPa , despite the fact that σ_c in this system is only 676 MPa . Strengths of this magnitude are rarely if ever seen in polycrystalline ceramics and are closer to those expected for single crystal fibers. Single crystal fibers, however, do not share the flaw tolerance capability that these laminates would.

Experimental Validation of the Model

[0035] To test the threshold strength concept and its predicted improvement of flaw tolerance, starting flaws of varying size were introduced into laminar ceramic composite specimens made up of tensile layers composed of aluminum oxide (Al_2O_3) and compressive layers composed of a mixture of mullite ($3 \text{ Al}_2\text{O}_3: 2 \text{ SiO}_2$) and Al_2O_3 . Residual compressive stresses of ~ 1.2 GPa were developed in the mullite/alumina layers during cooling from processing temperatures due to the mismatch of the thermal expansion coefficients of the tensile and compressive layers. These specimens were then tested to failure and the strengths were compared to those of unreinforced monolithic specimens with similar sized starting flaws.

[0036] Figure 3 shows that, as predicted, the failure stress of the laminates was relatively independent of the initial flaw size, while the strength of the monolithic alumina without the compressive layers was strongly flaw dependent. In addition, the morphology of the fracture surfaces of these specimens (Fig. 4) indicated that not only did the compressive layers arrest the initial propagation of otherwise catastrophic cracking initiating from the starting flaws, but they also allowed the specimens to endure a substantial amount of further loading before failure occurred, despite the presence of cracks that constituted a significant portion of the load-bearing cross-section of the specimen.

[0037] Further testing of laminate specimens with varying architectures [12] has validated our theory through independent investigation of the effects of the three most important independent variables in Eq. 3, namely the magnitude of the residual compression, the effect of the compressive layer thickness, and the effect of the tensile layer thickness. Figures 5, 6, and 7 show the effect of these parameters and their close correlation to the strengths predicted through the application of Eq. 3. However, the data also show that the current theory ceases to accurately describe the experimental data when the magnitude of the residual compression is large and/or when the thickness of the compressive layers is large.

[0038] Closer inspection of the propagation of the cracking within the compressive layers [13] has revealed that the reason for this discrepancy lies in the presence of two distinctly different crack propagation modes. As seen in Fig. 8,

cracking in laminates with thin compressive layers under moderate levels of residual compression is observed to propagate in a stable manner, straight across the compressive layers, exactly as was assumed in the derivation of the current theory. In these cases, close correlation is seen between the predicted and measured threshold strengths, as well as the predicted and measured propagation lengths, as seen in Fig. 9.

[0039] However, observations also show that once the residual compression and/or the compressive layer thickness exceed a critical value, edge cracking [14] appears on the surface, as seen in Fig. 10, which prevents the further propagation of the crack until failure occurs at much higher loads. Measured failure strengths in these cases generally far exceed those predicted by Eq. 3, while the arrest of the cracking at the edge crack conflicts with predictions of further extension, as seen in Fig. 11. Inspection of the fracture surfaces of these specimens (Fig. 12) and observation of the propagation of the crack beneath the penetration depth of the edge crack (Fig. 13) shows that bifurcation of the crack occurs, which begins to explain the discrepancies in measured and predicted threshold strengths. It is intuitively obvious that it will take much more stress to drive two cracks simultaneously at angles severely inclined to the applied tensile stress axis. Crack propagation of this type is not accounted for in the current model, which therefore invalidates its application in these cases.

Further Finite element Analysis

[0040] We are currently exploring the cause of bifurcation with the use of finite element analysis [15]. Early analysis has already shown that the single crack has a higher stress intensity factor relative to two bifurcating cracks. This would be expected since more energy is needed to drive two cracks than one. Thus, the reason does not lie within the magnitude of the strain energy released. Our current using the finite element approach to determine the T stress in front of the crack, as a function of the crack position within the compressive layer, has shown more success. The T stress is not dependent on either the radius vector or the angular position from the crack tip. When the T stress is compressive, the crack is expected to propagate on its symmetry plane; when it is tensile, the crack is expected to deviate onto another plane. Our finite element analysis does show that the T stress becomes a tensile stress

when the compressive stress exceeds a given value and therefore begins to explain the cause of bifurcation. Further exploration of this approach is in progress.

[0041] Further finite element simulations have also shown that the arrest phenomena are strongly affected by the moduli of the laminae [16]. As can be seen in Fig. 14, the value of the stress intensity factor is decreased as it enters a compressive layer with a lower modulus relative to the thicker, tensile layer. That is, a compressive layer with a lower elastic modulus will further increase the threshold strength because it stores less elastic strain energy relative to a compressive layer made with a material with a higher elastic modulus. Figure 14 also shows that if the elastic modulus of the compressive layer is too large, it may not stop crack extension at all. Thus, the most desired material couples that exhibit a threshold strength should have a compressive layer modulus less than the tensile layer modulus.

Developing Compressive Stresses via a Structural Phase Transformation

[0042] Although most of the experiments to test the initial model were performed with alumina and mullite/alumina layered materials that used the differential thermal contraction during cooling to develop the compressive stresses, we have also initiated a study [17] that uses a structural phase transformation to induce the biaxial compressive stresses. In the current case, we use the tetragonal to monoclinic phase transformation of unstabilized ZrO_2 to induce the compressive stresses. In this case, the thick (tensile) layers were composed of Al_2O_3 , and the thinner compressive layers were formed of a mixture of unstabilized ZrO_2 and Al_2O_3 . It was expected that the magnitude of compressive stresses would be controlled by the fraction of unstabilized ZrO_2 in the mixed layer, due to its molar volume increase during the structural phase transformation that occurs during cooling.

[0043] Figure 15 reports the threshold strength as a function of the fraction of unstabilized ZrO_2 in the mixed layer. As shown, the threshold strength is high, but does not change with the volume fraction of ZrO_2 for fractions greater than 0.30 volume fraction of the unstabilized ZrO_2 . This system is currently under study to understand the relation between the transformation and compressive stresses that arise during the biaxially constrained phase transformation.

Extension of the Threshold Concept to More Isotropic 3-Dimensional Architectures

[0044] While the majority of the work undertaken thus far has focused on 2-dimensional laminated materials, it is immediately apparent that these anisotropic architectures will be unable to yield a threshold strength in conditions other than the highly simplified ones described thus far (i.e. loading direction parallel to the laminar plane, driving cracks oriented normal to the layers). In order for the threshold concept to be more versatile and robust, higher dimensionality composite architectures must be developed that allow for more isotropic arrest behavior.

[0045] As shown in Figs. 16 and 17, we have discovered two different architectures where the compressive layer will stop cracks that extend in any direction. In the first [18], rods of one material are coated with a second material that will produce the compressive stresses. After bundling the coated rods, they are compressed and densified at high temperature to form a solid body that approximates an array of hexagonal prisms, with separated compressive layers on all sides as shown in cross section. Cracks that extend either along the axis or across the diameter of the hexagonal prisms will be stopped by the compressive layers that mutually bond the hexagonal prisms into a solid body.

[0046] In the second method [19], spheres of a material are coated with a second material that will form the compressive layer. As shown in Fig. 17, the spheres are mutually deformed by an applied isostatic pressure that converts the spheres into polyhedra that are separated by layers of a material that will form compressive layers. This array of polyhedra is then densified with a high temperature heat treatment to form a dense, solid body composed of polyhedra of one material surrounded and bonded together by compressive layers of a second material. It is obvious that cracks that extend in any direction within the material forming the polyhedra will be stopped by the compressive layers.

[0047] Although the foregoing invention has been described in some detail by way of illustration and example for purposes of clarity and understanding, various modifications and changes which are within the knowledge of those skilled in the art are considered to fall within the scope of the claims.

Supplemental Descriptive Material

[0048] The following list contains descriptive information that elaborates upon and clarifies the claims set forth below:

[0049] Clarification of Claim 1: All other factors being equal, the smaller the separation distance between compressive regions, the higher the threshold strength. The threshold strength is optimized when the distance separating the regions of the material(s) that do(es) not contain compressive stresses is between 0.2 and 0.01 times the dimension of the material(s) that do(es) not contain the compressive stresses, as measured from the interface between the materials. All else being equal, the larger the compressive stress, the larger the threshold stress. Compressive stresses in the range of 500 MPa to 5000 MPa are desired.

[0050] Clarification of Claim 2: The layers of materials containing biaxial, residual, compressive stresses are known as compressive layers.

[0051] Clarification of Claim 9: Examples of two materials chosen from this list would be alumina and zirconia, where the compressive stresses would arise in the alumina due to its lower thermal contraction coefficient; another example would be silicon nitride and silicon carbide, where the compressive stresses would arise in the silicon nitride due to its lower thermal contraction coefficient. A third example would be alumina and mullite, where the compressive stresses would arise in the mullite during cooling due to its lower thermal contraction coefficient.